Waste Foundry Sand Reused as Clay Replacement for Tile Manufacture

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ABSTRACT

In an effort to extend the reutilization of waste foundry sand (WFS), we evaluated the use of WFS as a replacement for clay in tile manufacturing. The effects of WFS addition on the properties of clay tile specimens were investigated at six different levels of WFS replacement: 0, 10, 15, 20, 40, and 60%. These specimens were fired at one of three different kiln temperatures: 1,000, 1,050, and 1,100ºC. Various physical analyses were performed to determine the properties of the finished tiles. The results of this study indicate that the properties of the tile specimens were improved by WFS supplementation and that the optimal clay replacement level was 15% WFS.

1. INTRODUCTION

The goals of reutilizing industrial by-products such as waste foundry sand (WFS) include reducing the use of natural resources and ameliorating the negative environmental and ecological impacts of industrial waste. Moreover, the reutilization of industrial waste can achieve the objective of sourcing green construction materials to reduce energy consumption and waste generation. The Taiwanese government allows WFS produced by industries such as the basic metal industry, the fabricated metal products industry, and the mechanical apparatus manufacturing and fixing industry to be directly reused. The main chemical components of these WFSs are silicon dioxide, aluminum oxide, and ferric oxide. These components are can improve the compactness and strength of reclaimed tiles (those manufactured from recycled or reused materials such as sewage sludge ash and WFS in admixture with clay).

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When WFS is reused as a construction material, it is typically employed as a raw material in cement or an aggregate in concrete. Siddiquea et al. [1] analyzed the physical and chemical properties of WFS and measured its leachate characteristics using various testing techniques, pointing out that WFS can be used to produce high-quality concrete, soil amendments, flowable fills, and embankments. The use of WFS helps to reduce waste disposal problems. Bhat and Lovell [2] employed WFS as a fine aggregate along with fly ash in the manufacture of controlled low-strength material (CLSM) and developed CLSM mixes designed specifically to replace natural aggregates and cement with WFS. Lin et al. [3] studied the optimal substitution percentages for producing cement from WFS including shell molding sand (7.6 wt.%) and sodium silicate sand (10.8 wt.%) at a kiln temperature of 1,400ºC and reported that it is feasible and safe to apply these materials in the cement manufacturing process. Reis and Jurumenh [4] investigated the fracture characteristics of polymer concrete made with WFS and noted that the use of WFS increased the fracture strength of polymer concrete, resulting in a final fracture energy similar to that of the standard formulation. The authors stated that WFS is an excellent alternative raw material. Park et al. [5] recycled WFS by applying regeneration (including comminution and separation) and stabilization processes, suggesting that it was possible for the foundry industry to replace "green" sand with regenerated sand.

Zanetti and Fiore [6] applied bentonite-bonded molding sands as binding agents for the production of tiles in the ceramic industry and concluded that fine particles improved the tile product and that the reclamation of bentonite molding sands conserved both raw materials and landfill space, providing both economic and environmental advantages. Alonso-Santurde et al. [7] developed three leaching tests to evaluate the environmental behavior of the ceramic bodies of both commercial and foundry-sand-based bricks in different stages of their life cycles. Although these bricks may cause environmental problems at the end-of-life stage, they suggested that foundry wastes can be used as raw materials in a viable, sustainable brick manufacturing process. Zheng et al. [8] employed WFS and other solid wastes in tile manufacturing and found that these tiles met the Chinese standards. Moreover, the heavy metal concentrations obtained from leachate tests were lower than those required by the standards. The reuse of WFS as a raw material can reduce the cost of tile manufacture. Shpector et al. [9] employed WFS in building brick manufacturing. After evaluating the bricks using leaching tests, the authors suggested that WFS can be reused as a construction material.

Studies related to the use of WFS in tile manufacturing have been less frequently reported in the literature, especially discussions regarding the amount of clay replaced with WFS and the kiln temperatures at which the tiles were fired. In general, the abrasion and bending strength of reclaimed tiles are lower than those of clay tiles. However, WFS contains a higher amount of Fe_2O_3 than other solid wastes (such as fly ash and sewage sludge ash), and this particular chemical component is the primary constituent of tile. In this study, we investigated the effects of WFS substitution rates and other conditions of tile manufacture such as kiln temperatures. We hope that the results of this study can extend the reutilization of WFS in construction materials.

2. EXPERIMENTAL METHODS
2.1. Materials

Clay and WFS are the two primary materials used to manufacture tile specimens in this study. Table 1 lists the chemical compositions of the clay and the WFS. The specific gravity of the clay was 2.08. The primary elemental constituents of the clay were Si (23.13%) and Al (15.65%), and the secondary constituents were K (7.42%) and Ca (4.83%), all present as oxide compounds, with the balance consisting of O (48.97%). The specific gravity of the WFS was 2.57, and the WFS powder was passed through a #100 sieve. As shown in Table 1, the primary elements comprising the WFS were Si (27.15%), C (7.65%), and Fe (7.21%), again as oxides, with the balance consisting of O (47.24%). Furthermore, the secondary chemical elements were Al (5.30%), Ca (2.35%), and Mg (1.19%). Table 2 presents the results obtained from a TCLP test performed on the WFS. The concentrations of all heavy metals met the requirements set by the Taiwanese EPA.

In this study, six levels of WFS replacement were evaluated: 0, 10, 15, 20, 40, and 60%. Reclaimed tile specimens were manufactured by uniformly mixing different quantities of WFS and clay with suitable amounts of water. Before the mixture was formed into strips, air was expelled from the mixture using a de-airing pug mill and a press machine. The strips were then cut into tiles of dimensions 12 cm × 6 cm × 1 cm. After a few days of development in a room with a temperature of approximately 27ºC, these reclaimed tile specimens were fired in an electric furnace at a controlled temperature of 1,000, 1,050, or 1,100ºC to complete the tile manufacturing process. The procedure for making reclaimed tile specimens is illustrated diagrammatically in Figure 1. After the specimens cooled, shrinkage, water absorption, weight loss on ignition, bending strength, and acid-alkali resistance tests were performed on the reclaimed tile specimens. The microstructures of the specimens were observed by X-ray and SEM. The parameters for each test are provided below.

Table 1. The chemical compositions of the clay and the foundry sand

Chemical elements (%)	O	C	Al	Si	S	Mg	K	Ca	Cr	Fe	Zn
Clay	48.97	—	15.65	23.13	—	—	7.42	4.83	—	—	—
Foundry Sand	47.24	7.65	5.30	27.50	0.06	1.19	0.67	2.35	—	7.21	0.85

Table 2. Results of toxicity characteristic leaching procedure (TCLP) test of the WFS

(mg/L)	As	Pb	Cu	Cd	Zn	Cr	Hg	Cr^{+6}	Se	Ba
Foundry sand	N.D	N.D	N.D	N.D	11.3	<0.2	N.D	N.D	N.D	<0.035
Taiwan TCLP regulatory limits	5.0	5.0	15.0	1.0	25.0	5.0	0.2	2.5	1.0	100.0

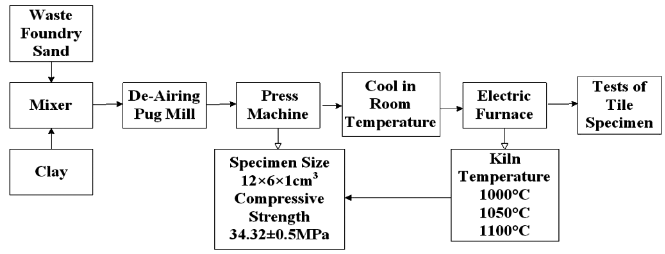


Figure 1. The procedure for making reclaimed tile specimens

2.2. Shrinkage Test

The method of measuring the drying and firing shrinkage of reclaimed tile specimens is regulated by CNS 2887. Tile shrinkage is defined as the ratio of the lengths of the specimen before and after firing.

2.3. Weight Loss on Ignition

The method of measuring the weight loss on ignition of reclaimed tile specimens is regulated by CNS3299. Weight loss on ignition measures the difference in the weights of each tile before and after firing. This weight loss is primarily due to the evaporation of moisture, the oxidative decomposition of organic matter, the decomposition of inorganic salts, and trace emission of heavy metals at high temperature.

2.4. Water Absorption

The method of measuring the water absorption of reclaimed tile specimens is regulated by CNS3299. For the water absorption test, specimens are oven-dried at 105 to 110ºC for 3 hours and then placed in a desiccator to cool to room temperature. The specimens are weighed immediately after cooling. The specimens are then submerged in water for 24 hours, removed, patted dry with a lint-free cloth, and weighed. The water absorption is defined as the ratio of the weight of the water absorbed by the specimens to the weight of the dry specimens.

2.5. Bending Strength

The bending strength test was performed on the reclaimed tile specimens according to the CNS3299 standards. The bending strengths of the specimens are affected by the porosity, pore distribution, and vitrification of the tile bodies and their crystal morphologies.

2.6. Acid-Alkali Resistance

The acid-alkali test was also completed according to the CNS3299 standards. In this test, samples are cleaned with liquid detergent, placed in an oven at 105ºC for 3 hours, and then cooled to room temperature before being exposed to the test solutions.

1. Acid test: samples are placed in a hydrochloric acid solution for 24 hours and then rinsed under running water to remove any residual acid solution. The samples are

examined for differences in appearance, including color changes and other abnormal reactions. If a sample is visibly attacked, the results of the acid test are recorded as "affected."

2. Alkali test: samples are placed in a potassium hydroxide solution for 24 hours and then rinsed under running water to remove any residual alkali solution. The samples are examined for differences in appearance, including color changes and other abnormal reactions. If a sample is visibly attacked, the results of the alkali test are recorded as "affected."

2.7. Field-Emission Scanning Electron Microscopy (FE-SEM)
A high-resolution Hitachi S-4700 SEM (manufactured in Japan) was used to observe the surface microstructures of samples and small pieces of reclaimed tile bodies. The samples were coated with gold for better conductivity. The sample surfaces were examined at magnifications of 5,000X to obtain the microstructure images.

2.8. X-Ray Diffractometry (XRD)
A Shimadzu XRD-6000 X-ray diffractometer was used to characterize test samples that were ground to a powder and passed through a #100 sieve. Before the X-ray analysis, the powder was dried in an oven at 105ºC for 24 hours and then placed in a desiccator to cool. After testing, the results were compared with data in the Joint Committee on Powder Diffraction Standards (JCPDS) database to determine the major crystalline components of the tile samples. The test parameters were a scan rate of 2º/min, a voltage of 40 kV, a current of 30 mA, and a 2θ range of 5º to 60º.

3. RESULTS AND DISCUSSION
3.1. Shrinkage Test
Shrinkage is one of the primary phenomena observed upon firing a tile body and reflects possible increases in the numbers of contact points or surfaces among the particles and an increase or decrease of the number of pores within the tile body. Figure 2 presents the results of shrinkage tests performed on reclaimed tile specimens containing various levels of WFS and fired at various kiln temperatures. The figure shows that the shrinkage of the specimens decreased as the WFS substitution rate increased at 1,000ºC, implying that particles in the tile body were effectively integrated by the heating during the firing process. Similar results were observed at 1,100ºC, at which the shrinkages were 7.39, 6.85, 5.721, and 4.14% at WFS replacement levels of 15, 20, 40, and 60%, respectively. This pattern indicates that increasing the level of WFS replacement reduces the shrinkage of reclaimed tiles.

3.2. Weight Loss on Ignition Test
Figure 3 provides the results of the weight loss on ignition tests performed on reclaimed tile specimens containing various levels of WFS and fired at various kiln temperatures. The weight loss on ignition was between 5 and 9% for the specimens fired at 1,000ºC. The weight loss on ignition was apparently reduced at replacement rates greater than 20%, indicating that the weight loss decreased as the WFS substitution rate increased.

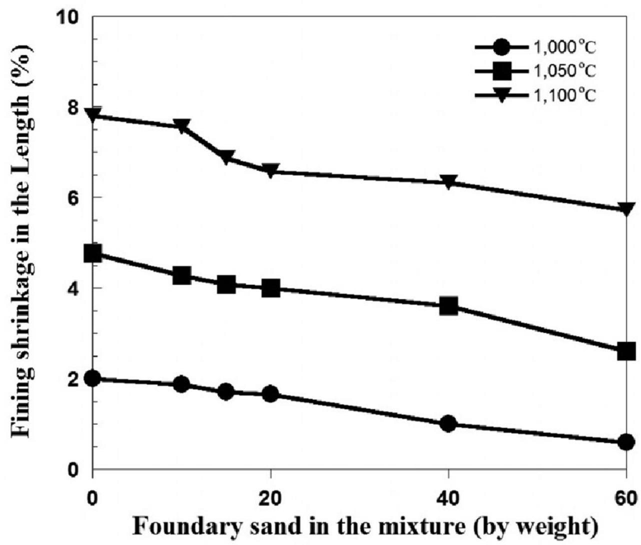


Figure 2. The results of shrinkage tests performed on reclaimed tile specimens containing various levels of WFS and fired at various kiln temperatures

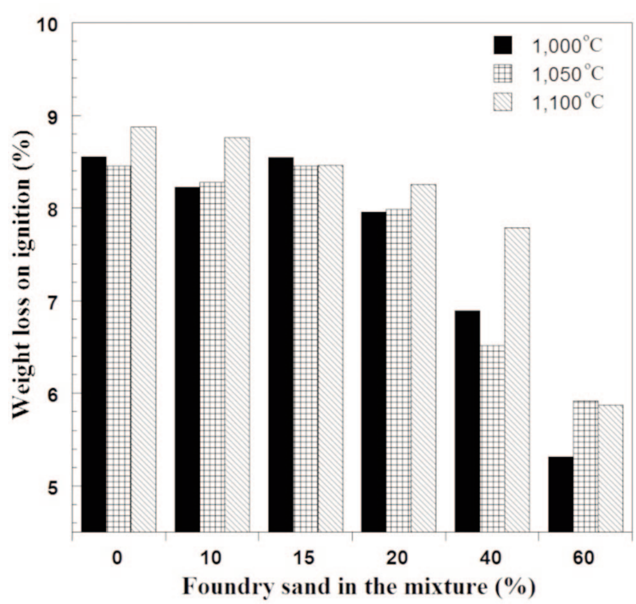


Figure 3. The results of weight loss on ignition tests performed on reclaimed tile specimens containing various levels of WFS and fired at various kiln temperatures

Moreover, the lowest weight loss on ignition was observed for specimens with 60% WFS replacement. This reduction in the weight loss on ignition was partly due to the larger particle size of WFS compared to clay. Furthermore, due to the higher iron content of the WFS, the binding interactions among particles may not be fully developed during the firing process. As a result, the reclaimed tile specimens exhibited less weight loss on ignition at higher WFS contents.

3.3. Water Absorption Test

Figure 4 shows the results of water absorption tests performed on reclaimed tile specimens containing various levels of WFS and fired at the abovementioned kiln temperatures. Due to the larger particle size of the WFS, pores of different sizes were distributed throughout the bodies of the reclaimed tile samples. Hence, the interior of the tile body was not as smooth and dense as the clay tile surface, which exhibited better water resistance. Moreover, these pores were sometimes connected to form passageways, allowing moisture to more easily intrude into the reclaimed tile body. As a result, the water absorption rates of reclaimed tile specimens are higher than the controls (tile specimens with no WFS replacement), as shown in Figure 4.

Figure 4 also demonstrates that the water absorption rate of the control group was approximately 18.73% when the kiln temperature was at 1,000ºC. The water absorption values for the specimens with 10, 15, 20, 40, and 60% WFS replacement levels were 19.62, 19.77, 20.87, 26.89, and 30.48%, respectively. As stated above, the replacement of clay with WFS in these specimens increased the internal porosity of the tile body.

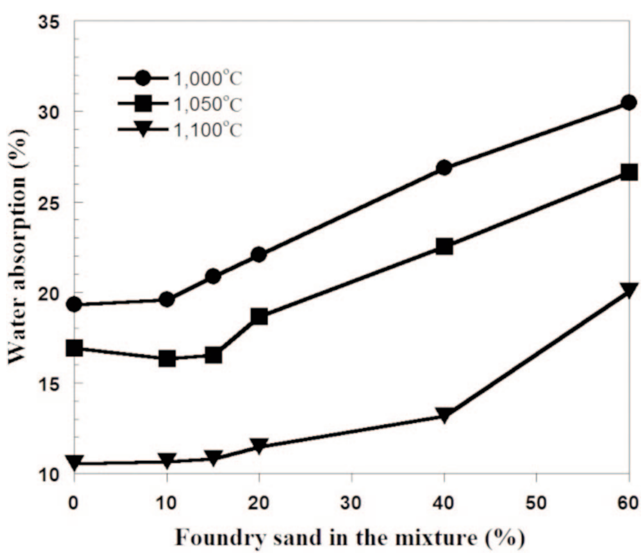


Figure 4. The results of water absorption tests performed on reclaimed tile specimens containing various levels of WFS and fired at various kiln temperatures

This increase in porosity led to higher water absorption values and was readily apparent at WFS replacement levels greater than 40%.

3.4. Bending Strength Test

Figure 5 displays the results of bending strength tests performed on reclaimed tile specimens produced with various levels of WFS replacement and fired at the above mentioned kiln temperatures. As shown here, the bending strengths of the specimens with 15% WFS replacement were higher than those of the control group. The increases in bending strength were 1.29 MPa at 1,000ºC and 3.39 MPa at 1,100ºC. This finding suggests that the Fe_2O_3 in the WFS improved the strength of the specimens. Furthermore, the bending strengths of the specimens increased as the WFS content increased up to 15%. However, when the replacement rate increased beyond 15%, the strength began to decrease, particularly rapidly for specimens containing over 40% WFS.

3.5. Acid-Alkali Resistance Test

Figure 6 depicts the weight losses for the reclaimed tile specimens containing 0, 15, and 40% WFS replacement and fired at a kiln temperature of 1,000ºC after soaking in acid and alkali solutions for 1, 3, 7, and 14 days. The weight losses of the specimens increased as the soaking time increased in both solutions. The weight losses observed after soaking in the alkali solution were less than those in the acid solution. Hence, the specimens were more resistant to alkaline corrosion than to acidic corrosion.

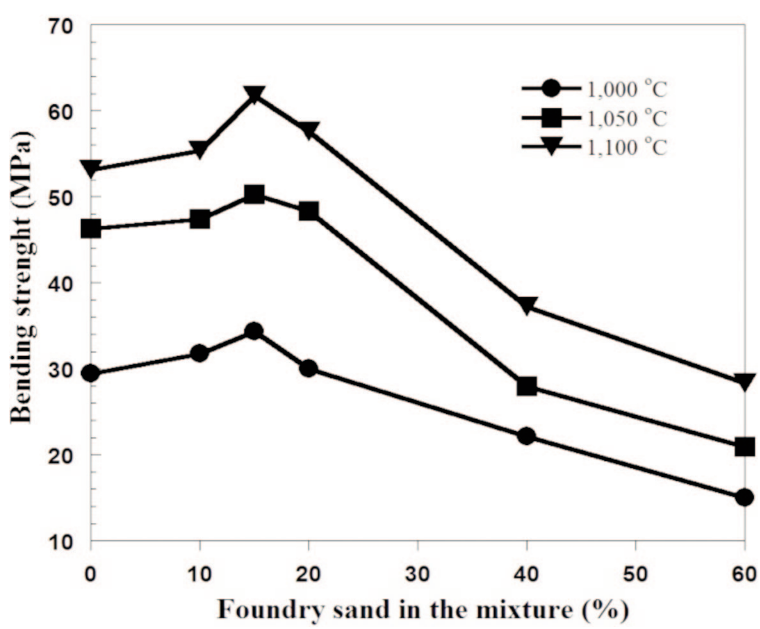
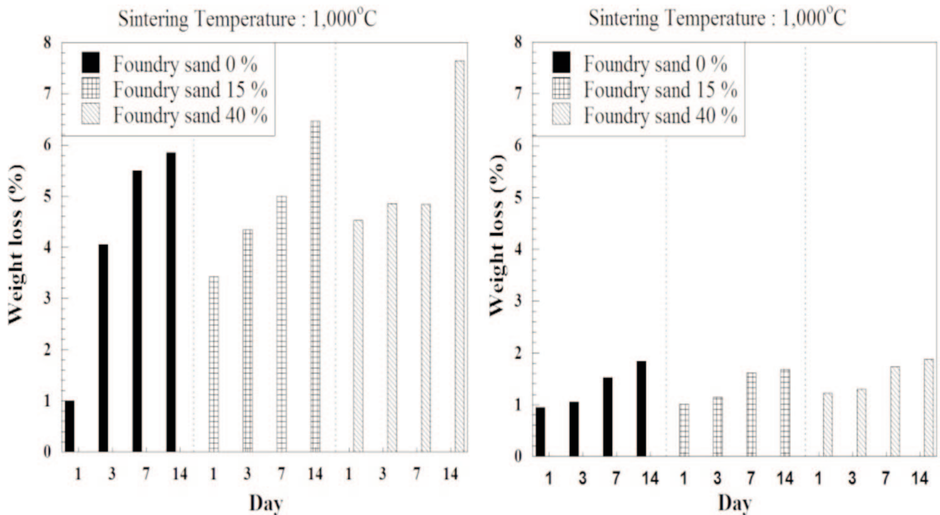
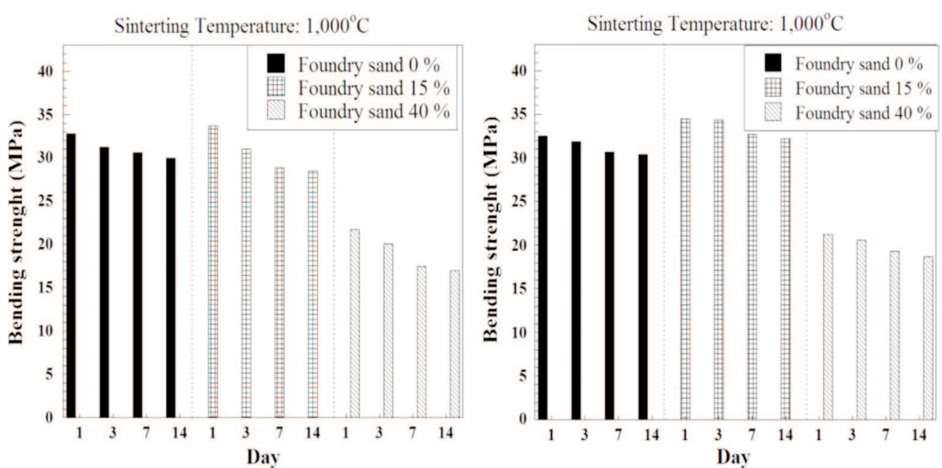


Figure 5. The results of Bending Strength tests performed on reclaimed tile specimens containing various levels of WFS and fired at various kiln temperatures



(a) in acid solution (b) in alkali solution

Figure 6. The weight losses for the reclaimed tile specimens containing various WFS replacement and fired at a kiln Temperature of 1,000ºC after soaking in acid and alkali solutions for 1, 3, 7, and 14 days



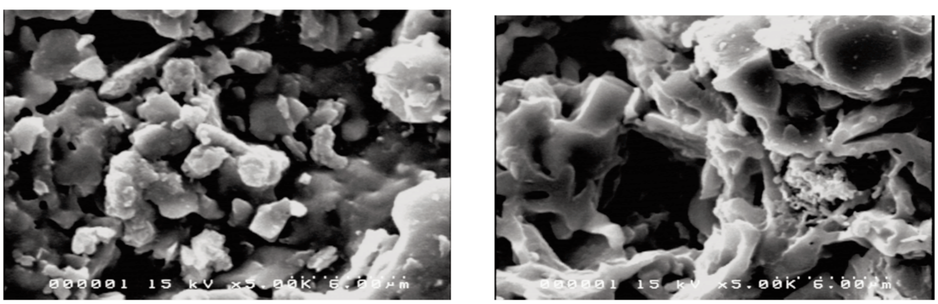
(a) in acid solution (b) in alkali solution

Figure 7. The bending strength for the reclaimed tile specimens containing various WFS replacement and fired at a kiln Temperature of 1,000ºC after soaking in acid and alkali solutions for 1, 3, 7, and 14 days

3.6. SEM Analysis

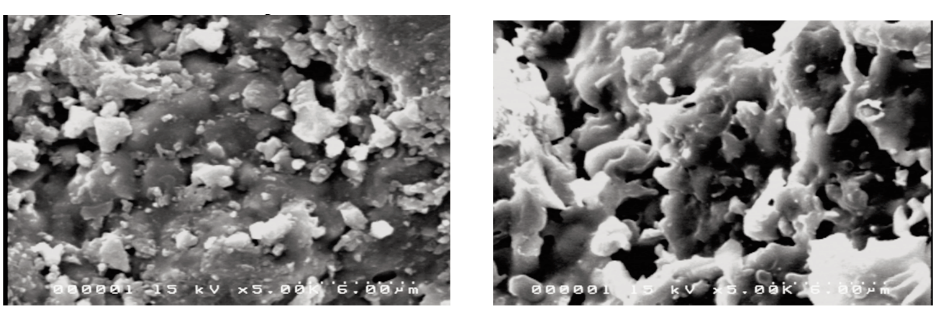
To understand the effects of WFS replacement on the characteristics of the reclaimed tile specimens, an SEM analysis was performed to observe the changes in the microstructures of the tile bodies, e.g., the crystalline states and porosities. The samples were small pieces from the surfaces and interiors of the tile bodies and were examined at 5,000X to obtain microstructure images. Figures 8 and 9 present SEM images of the specimens produced with 0 and 15% WFS replacement and fired at a kiln temperature of 1,000ºC.

Heat transfer occurred in the interior of the tile body due to the high temperature of the firing process. When the kiln temperature reached 1,000ºC, the particles bonded to each other. Figure 8, a 5,000X micrograph of a sample from the control group, depicts a necking effect on the surface of the specimen. Hence, pores of different sizes were distributed on the tile surface. Moreover, the heat was acted as a driving force for the diffusion of particles in the interior of the tile body. Consequently, diffusion bonding resulted in the formation of crystalline structures in the interior of the tile body. However, the pore sizes were large and irregularly shaped. Figure 9 shows that when



(a) 5,000X (Surface) (b) 5,000X (Interior)

Figure 8. SEM images of the specimens produced with 0% WFS replacement and fired at a kiln temperature of 1,000ºC



(a) 5,000X (Surface) (b) 5,000X (Interior)

Figure 9. SEM images of the specimens produced with 15% WFS replacement and fired at a kiln temperature of 1,000ºC

15% of the clay was replaced with WFS, the surface particles were well bonded to each other and the pore sizes were small. Moreover, the bonded surfaces were relatively flat, and the pore channels were smooth. In the interior of the tile body, necks were gradually formed, resulting in better adhesion interactions among the particles. The diffusion bonding was better than that of the control group. This result suggests that the application of WFS in tile manufacturing is viable and can improve the compactness of the tile structure.

3.7. XRD Analysis

Figure 10 shows the XRD results for reclaimed tile specimens produced with 0, 15, 20, 40, and 60% WFS replacement levels before and after firing at a kiln temperature of 1,000ºC. Changes in the peak intensities of the various mineral components of the specimens with different WFS contents were less apparent before firing than after firing. However, the peak intensities of both SiO_2 and Fe_2O_3 at $2\theta = 50.32^o$ and $2\theta = 36.70^o$, respectively, increased slightly as the amount of WFS increased. After firing at 1,000ºC, the peak intensity of SiO_2 at $2\theta = 50.3^o$ increased as the WFS content increased from 0 to 15% and then decreased slightly as the WFS content increased from 15 to 60%. Hence, the highest SiO_2 peak intensity was observed for the tile specimen with a WFS content of 15%. The reclaimed tile specimens exhibited better siliconization than the control group. Hence, the specimens with WFS replacement exhibited improved compactness, especially the specimens with 15% WFS replacement. This result is confirmed by the observations of the SEM analysis (see Figures 8(a) and 9(a)). The 15% WFS specimens exhibited the improvement on pore sizes and open structures on their surfaces. The surface particles were well bonded with each other, and the surface structure was compact. This study suggests that a WFS replacement level of 15% improved the siliconization of the specimens.

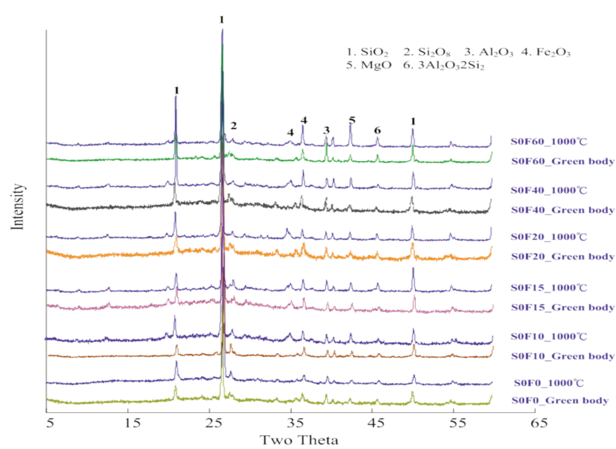


Figure 10. The XRD results for reclaimed tile specimens produced with 0, 15, 20, 40, and 60% WFS replacement levels before and after firing at a kiln temperature of 1,000ºC

As stated above, the siliconization of tile is related to the intensity of the SiO_2 peak. Figure 10 displays the greater degree of siliconization observed at $2\theta = 26.74^{\circ}$ after firing at 1,000ºC. Similar results were observed in the 2θ range from 20 to 20.9º. With 15% WFS replacement, the peak intensity of mullite became relatively higher after firing at 1,000ºC, while Fe_2O_3 was less affected by the firing process. Moreover, at $2\theta = 39.64^{\circ}$, when the WFS content was less than 20%, the Al_2O_3 peak intensities after firing at 1,000ºC were higher than those before firing. However, contrary results were obtained at WFS contents greater than 20%. Because the amount of Al_2O_3 affected the crystallization of the tile body, the above observation implies that when the level of WFS replacement was greater than 20%, the crystalline state of the tile body was not well developed after firing at 1,000ºC.

4. CONCLUSIONS

In this study, waste foundry sand was utilized as a replacement for clay in tile specimen manufacturing. After evaluating the tiles, we drew the following conclusions:

1. The shrinkages of the tile specimens after firing at 1,000ºC decreased as the WFS content increased, implying that particles in the tile body were effectively integrated by the driving force of the heat applied during the firing process. This result suggests that increasing the rate of waste foundry sand replacement can reduce the shrinkage values of tile specimens.
2. The results of the weight loss on ignition test indicate that the weight losses of the tile specimens decreased as the WFS content increased. Moreover, tile specimens with waste foundry sand substitution exhibited less weight loss after firing at each of the tested firing temperatures.
3. Because the waste foundry sand has a larger particle size than clay, pores of different sizes were distributed throughout the tile body in specimens with waste foundry sand supplementation. This increase in porosity led to greater water absorption, which became readily apparent at foundry sand replacement rates greater than 40%.
4. The bending strengths of the tile specimens increased as the amount of waste foundry sand increased up to 15%. As determined from SEM images, when clay was replaced with 15% waste foundry sand, the surface particles were well bonded with each other and the pore sizes were small. Moreover, the bonded surfaces were relatively flat, and the pore channels were smooth. In the interior of the tile body, necks were formed, resulting in improved adhesion interactions among the particles. The diffusion bonding was better than that of the control clay tile specimens. However, when the WFS replacement level increased beyond 15%, the bending strength was reduced and rapidly decreased for tile specimens with more than 40% waste foundry sand replacement.
5. After soaking in both acid and alkali solutions, the strengths of the specimens with 15% waste foundry sand replacement were greater than those of specimens with 0 and 40% replacements. This result indicates that specimens with 15% WFS replacement exhibited the best resistance to acid and alkali attack. However, higher

amounts of waste foundry sand replacement may negatively affect the resistance of tile specimens to acid and alkali attack.

6. The SiO_2 peak intensity is related to the siliconization of the tile specimens. After firing at a kiln temperature of 1,000ºC, the SiO_2 peak intensity at $2\theta = 50.3^{\circ}$ increased as the amount of WFS replacement increased from 0 to 15%, and the highest SiO_2 peak intensity was observed for the tile specimen with 15% WFS replacement. Hence, tile specimens produced with WFS replacement exhibited better siliconization than the clay tile specimens.

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